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IN THE UNITED STATES PATENT & TRADEMARK OFFICE

IN RE APPLICATION OF : :

Heiko MAAS ET AL : EXAMINER: DANG, T. D.

SERIAL NO: 09/914,532 : :

FILED: AUGUST 30, 2001 : GROUP ART UNIT: 1764

FOR: OLIGOMERIZATION OF C6-OLEFINS

APPEAL BRIEF

COMMISSIONER FOR PATENTS  
ALEXANDRIA, VA 22313

SIR:

This is an appeal of the Office Action dated May 19, 2003, of at least twice rejected Claims 1-3 and 6-9. A Notice of Appeal is **submitted herewith**.

I. REAL PARTY IN INTEREST

The real party in interest in this appeal is BASF Aktiengesellschaft having an address at 67056 Ludwigshafen, Germany.

II. RELATED APPEALS AND INTERFERENCES

Appellants, Appellants' legal representative and the assignee are aware of no appeals or interferences which will directly affect or be directly affected by or have a bearing on the Board's decision in this appeal.

### III. STATUS OF THE CLAIMS

Claims 1-4 and 6-9, all the claims in the application, stand rejected and are herein appealed.

### IV. STATUS OF THE AMENDMENTS

An amendment is **submitted herewith**.

### V. SUMMARY OF THE INVENTION

As recited in independent Claim 1, the invention is a process for oligomerizing C<sub>6</sub>-olefins by reaction of a C<sub>6</sub>-olefin-containing reaction mixture over a nickel-containing fixed-bed catalyst, comprising from 10 to 70% by weight of nickel oxide, from 5 to 30% by weight of titanium dioxide and or zirconium dioxide and from 0 to 20% by weight of aluminum oxide as significant active constituents and silicon dioxide as the remainder, wherein the reaction over the fixed-bed catalyst is carried out continuously in the liquid phase and run at a conversion to oligomerized C<sub>6</sub>-olefins of from 10 to 30% by weight based on a throughput of the reaction mixture through the catalyst in a single pass.

See original Claims 1, 2 and 5, and the specification at page 1, lines 32-36, and page 2, lines 1-4, 9-11 and 24-25.

### VI. ISSUES

(A) Whether Claims 1-4 and 6-8 are unpatentable under 35 U.S.C. §103(a) over U.S. 5,849,972 (Vicari et al) in view of U.S. 5,243,118 (Sanderson et al)?

(B) Whether Claim 9 is unpatentable under 35 U.S.C. §103(a) over Vicari et al in view of Sanderson et al, further in view of EP395857A (Huls)?

(C) Whether Claims 6-9 are unpatentable under 35 U.S.C. §112, second paragraph?

### VII. GROUPING OF THE CLAIMS

For Issue (A) above, Claims 2, 3 and 6-8 each stand or fall separately from Claim 1.

For Issue (C) above, the claims each stand or fall separately.

### VIII. ARGUMENT

#### Issue (A)

Claims 1-4 and 6-8 stand rejected under 35 U.S.C. §103(a) as unpatentable over Vicari et al in view of Sanderson et al. That rejection is untenable and should not be sustained.

The present invention relates to a process for oligomerizing C<sub>6</sub>-olefins, in particular for preparing C<sub>12</sub>-olefins by dimerization. As described in the specification beginning at page 1, line 8, processes for the oligomerization of olefins are known, such as that disclosed in DE-A-4339713, which is the German equivalent of Vicari et al. As described in the specification herein at page 1, lines 9-14, Vicari et al describes a process for oligomerizing olefins to give highly linear oligomers. In this process, C<sub>2,6</sub>-olefins are reacted at superatmospheric pressure and elevated temperature over a fixed-bed catalyst comprising from 10 to 70% by weight of nickel oxide, from 5 to 30% by weight of titanium dioxide and/or zirconium dioxide, from 0 to 20% by weight of aluminum oxide as significant active constituents and silicon dioxide as the remainder.

As described in the specification at page 1, line 25, a disadvantage of the known processes is that the catalyst life is frequently too short. The catalyst is, in particular, clogged by higher oligomers and therefore loses its activity.

Applicants have discovered that by oligomerizing C<sub>6</sub>-olefins using the above-discussed fixed-bed catalyst of Vicari et al., at a conversion to oligomerized C<sub>6</sub>-olefins of not more than 30% by weight (now from 10 to 30% by weight), based on the reaction mixture, in a single pass, deactivation of the catalyst can be avoided and dimer selectivity can be increased. By recycling unreacted reaction mixture after separating off the oligomers, a higher total conversion can be obtained. Indeed, the presently-claimed process makes it possible to realize a total conversion of over 90% together with a C<sub>12</sub> (dimer) selectivity of over 80%.

Adherence to the conversion specified according to the claims greatly increases the operating life of the catalyst, since the formation of high-boiling compounds which can deposit on the catalyst and thus cause a drop in activity is suppressed, as described in the specification at page 2, lines 22-27.

Examples 1 and 2 in the specification illustrate the presently-claimed invention, and the above-discussed superior results. In Example 1, beginning with a hydrocarbon mixture feed containing 73% by weight of C<sub>6</sub> component, a C<sub>6</sub>-olefin conversion of 94.7% and a C<sub>12</sub> selectivity of 83.6%, based on the C<sub>6</sub>-olefins reacted, was obtained.

In Example 2, the C<sub>6</sub> concentration in the hydrocarbon mixture feed was 98.7% by weight. The result was a C<sub>6</sub>-olefin conversion of 98.4% and a C<sub>12</sub> selectivity of 82.6%, based on the C<sub>6</sub>-olefins reacted.

The Examiner has previously queried how the results in Examples 1 and 2 square with the above-discussed limitation of a maximum conversion of not more than 30% by weight. Applicants' attorney and the assignee's representative explained, referring to Figs. 1A or 1B, that the 30% conversion is with respect to the input to, and output from, reactor C1, while the final conversion and selectivity of the Examples is with respect to bottoms stream S. The

conversion regarding input to and output from C1 for each Example was calculated and is 20.2% for Example 1, and 19.4% for Example 2, as disclosed in the specification in the "Total C<sub>6</sub>," column and "Feed mixture to K1=reactor output" row, at the top of page 9, and the bottom of page 10, respectively.

The Heidemann Declaration of record provides a further demonstration of increased catalyst life. (The Heidemann Declaration indicates that attached thereto is an Experimental Report. Since counsel was not sure if the Report was actually attached when the Declaration was filed, a subsequent filing of the Report *per se* was made. Counsel represents that the respective Experimental Reports are one and the same.)

The catalyst used in the Experimental Report was the same catalyst employed in Examples 1 and 2 of the specification herein, as disclosed at page 7, lines 11-14. This catalyst was employed in each of Examples 1-3 of the Experimental Report.<sup>1</sup> In the Experimental Report examples, Example 1, the reaction temperature was 80°C; in Example 2, the reaction temperature was 160°C; and in Example 3, the reaction temperature was 60°C. The reaction pressure was 15 bars in all examples. Example 1 of the Experimental Report shows that conversion, based on a single path, was around 17%, even after 69 hours. In Example 3, it was around 20%, even after 168 hours. In Example 2, on the other hand, which was for purposes of comparison, the conversion steadily decreased, indicating catalyst deactivation, or stated in other words, a short catalyst life.

While Vicari et al discloses the presently-recited fixed-bed catalyst, as discussed above, Vicari et al do not recognize the nexus between maximum conversion in a single pass,

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<sup>1</sup>The Board should note that Examples 1-3 of the Experimental Report are different from Examples 1 and 2 described in the specification, although the same catalyst is used in each.

on the one hand, and increased catalyst life, higher selectivity of dimer, and higher overall conversion, on the other hand. Indeed, based on the examples and comparative examples in Vicari et al., the conversion varies from 41-55% for the comparative examples, and 53-78% for the examples. See the various tables at column 7 therein. Since no recycle is disclosed, it is assumed that these conversions are ultimate conversions. While this data is for C<sub>4</sub>-olefins, nevertheless, it is still sufficient to show that Vicari et al. do not recognize the significance of maintaining the conversion below a certain maximum for each single pass over the catalyst.

Sanderson et al. does not remedy any of the above-discussed deficiencies of Vicari et al. Sanderson et al. discloses a process for oligomerizing olefins using sulfate-activated molecular sieves. Sanderson disclose that by the use of such molecular sieves, a high percentage of dimers, i.e., a high dimer to trimer ratio, can be achieved (column 2, lines 35-37). Sanderson et al further disclose that the oligomerization results in an oligomer product distribution that varies with reaction time; as the reaction time increases, the olefin monomer conversion increases, and the selectivities for the heavier oligomers increase (column 3, lines 33-55). The olefin monomer feed stocks of Sanderson et al must contain at least 10 carbon atoms (column 3, lines 13-21).

In Sanderson et al., no particular olefin conversion goal is disclosed, although it is noted that liquid hourly space velocity may be varied within wide limits (column 5, lines 55-61), and it is noted that the olefin conversion, as specified in the table bridging columns 7 and 8, varies widely, from 3.0% to 70.4%. Moreover, the data therein show that conversion is not strictly a function of reaction time, as can be seen by comparing examples using the same catalyst. For example, Catalyst 4 produces a conversion of 3% in 5 hours (Example 1), and a conversion of 11.3% in 4 hours, but at a higher temperature (Example 7). A comparison of Examples 2 and 6; Examples 4 and 5; produces a similar result. Thus, the conversion was

actually lower after a longer reaction time. In addition, all the examples therein employed a reaction time of 4 or 5 hours, yet no trend can be discerned therefrom that conversion must be limited to 30% or lower to maximize dimer product. Rather, the trend is due to the use of sulfate-activated molecular sieves (Catalysts 2-4 and 7) compared to the non-sulfate-activated molecular sieves (Catalysts 1, 5 and 6). Compare Examples 3 and 5, where at identical feeds, reaction times and temperatures, the sulfated molecular sieve produced a conversion of 32.5% (Catalyst 2), while the same catalyst in non-sulfated form produced a conversion of 18.8% (Catalyst 1). Compare also Examples 11 and 13, which also shows increased conversion using the sulfated molecular sieve.

Sanderson et al adds nothing to Vicari et al. Sanderson et al is concerned with a different catalyst therefrom, and olefins having a carbon range outside the range specified by Vicari et al. Sanderson et al disclose nothing with regard to any possibility of increased catalyst life by limiting conversion percentage to a particular amount. Nor would one skilled in the art even combine Vicari et al and Sanderson et al in view of the mutually exclusive catalysts and olefins used. Indeed, without the present disclosure as a guide, one skilled in the art would not be motivated to combine Vicari et al and Sanderson et al. Even if combined, the result would not be the presently-claimed invention.

Thus, the combination of Vicari et al and Sanderson et al does not present a *prima facie* case of obviousness. However, even if such a case had been made out, the conversions of around 17% and around 20% demonstrated are sufficient to demonstrate patentability of the presently-recited 10 to 30% by weight conversion.

The Examiner's finding that Applicants must compare their process for all possible conversions within the claims, i.e., from above 0 to 30% by weight (now 10 to 30% by weight), to processes wherein the conversion ranges from 30% up to 100%, is clearly

erroneous. Since the prior art neither discloses nor suggests any nexus between conversion and other properties, such as catalyst life, Applicants were under no burden to provide **any** comparative data.

The Examiner criticizes the data as not being commensurate in scope with the claims. In reply, data of patentability must be commensurate in scope with only that part of claimed subject matter that is *prima facie* obvious. However, as discussed above, since the prior art does not recognize any nexus between conversion and catalyst life, there is **no** *prima facie* case of obviousness. Moreover, even if there was such a case, the Examiner may not consider such evidence only for its “knockdown ability”. As stated in *In re Carleton*, 599 F.2d 1021, 1024, 202 USPQ 165, 168-69 (CCPA 1979):

If the applicant presents rebuttal evidence, the decision-maker must consider all of the evidence of record (both that supporting and that rebutting the *prima facie* case) in determining whether the subject matter as a whole would have been obvious. [footnote omitted] *In re Rinehart*, 531 F.2d 1048, 189 USPQ 143 (CCPA 1976); *In re Lewis*, 443 F.2d 389, 170 USPQ 84 (CCPA 1971). The correct procedure for considering rebuttal evidence was set forth by this court in *In re Rinehart*, *supra* at 1052, 189 USPQ at 147:

Though the burden of going forward to rebut the *prima facie* case remains with the applicant, the question of whether that burden has been successfully carried requires that the entire path to decision be retraced. An earlier decision should not, as it was here, be considered as set in concrete, and applicant's rebuttal evidence then be evaluated only on its knockdown ability. Analytical fixation on an earlier decision can tend to provide that decision with an undeservedly broadened umbrella effect. *Prima facie* obviousness is a legal conclusion, not a fact. Facts established by rebuttal evidence must be evaluated along with the facts on which the earlier conclusion was reached, not against the conclusion itself. Though the tribunal must begin anew, a final finding of obviousness may of course be reached, but such finding will rest upon evaluation of all facts in evidence, uninfluenced by any earlier conclusion reached by an earlier board upon a different record.

The Examiner has found that Applicants' argument that by recycling unreacted mixture after separating off the oligomers, a higher total conversion can be obtained, is not persuasive. The Examiner's reason is that Applicants do not claim this feature. In reply, the Examiner is incorrect. This feature is claimed in Claim 8.

The Examiner has, in effect, dismissed all of the above arguments with regard to Sanderson et al by simply focusing on the disclosure at column 3, lines 33-55 thereof, concluding therefrom that one skilled in the art would vary conversion to optimize selectivity of oligomers. In reply, it is respectfully submitted that the Examiner has grossly simplified the disclosure in Sanderson et al. At any rate, as discussed above, Sanderson et al discloses no nexus between conversion in a single pass and selectivity, and clearly discloses and suggests no nexus between such conversion and catalyst life.

Claim 2 is separately patentable because the combination of Vicari et al and Sanderson et al neither discloses nor suggests the process as claimed in claim 1, wherein the reaction over the fixed-bed catalyst is run at a conversion to oligomerized C<sub>6</sub>-olefins from 10 to 25% by weight, based on the reaction mixture. Indeed, as discussed above, single pass conversions disclosed in the prior art appear to be much higher.

Claim 3 is separately patentable because the combination of Vicari et al and Sanderson et al neither discloses nor suggests the process as claimed in claim 1, wherein the oligomerization is essentially a dimerization. While dimerization is disclosed in the prior art, the oligomerization therein is not essentially a dimerization.

Claim 6 is separately patentable because the combination of Vicari et al and Sanderson et al neither discloses nor suggests the process as claimed in claim 1 which is carried out adiabatically in a shaft oven and in which part of the reaction mixture is returned to the reaction. The Examiner incorrectly finds that apparatus limitations are given no

patentable weight in method claims. The limitations in Claim 6 are limitations on the claimed method, and are not disclosed or suggested by the applied prior art.

Claim 7 is separately patentable because the combination of Vicari et al and Sanderson et al neither discloses nor suggests the process as claimed in claim 1, wherein a feed mixture is fractionated in a column to separate C<sub>6</sub>-olefins and oligomers prior to the reaction, the C<sub>6</sub>-olefins are returned to the reaction, the reaction mixture is returned to the column and the oligomers (C<sub>6</sub>-hydrocarbons) are discharged.

Claim 8 is separately patentable because the combination of Vicari et al and Sanderson et al neither discloses nor suggests the process as claimed in claim 1, wherein the reaction mixture after the reaction is fractionated in a column to separate C<sub>6</sub>-olefins and oligomers, the C<sub>6</sub>-olefins are returned to the reaction and the oligomers are discharged.

For all the above reasons, it is respectfully requested that the rejection over Vicari et al in view of Sanderson et al be REVERSED.

#### Issue (B)

Claim 9 stand rejected under 35 U.S.C. § 103(a) as unpatentable over Vicari et al in view of Sanderson et al, and further in view of Huls. That rejection is untenable and should not be sustained.

The Examiner relies on Huls for its disclosure of removing catalyst-poisoning components from a hydrocarbon stream. But, even if Huls were combined with Vicari et al and Sanderson et al, it would still not remedy the above-discussed deficiencies in the combination of Vicari et al and Sanderson et al.

Accordingly, it is respectfully requested that the rejection over Vicari et al in view of Sanderson et al and Huls be REVERSED.

Issue (C)

Claims 6-9 stand rejected under 35 U.S.C. § 112, second paragraph. That rejection is untenable and should not be sustained, for reasons stated in the amendment **submitted herewith.**

Accordingly, it is respectfully requested that this rejection be REVERSED.

IX. CONCLUSION

For the above reasons, it is respectfully requested that all the rejections still pending in the Final Office Action be REVERSED.

Respectfully submitted,

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## APPENDIX

### CLAIMS ON APPEAL

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Claim 1: A process for oligomerizing  $C_n$ -olefins by reaction of a  $C_n$ -olefin-containing reaction mixture over a nickel-containing fixed-bed catalyst, comprising from 10 to 70% by weight of nickel oxide, from 5 to 30% by weight of titanium dioxide and/or zirconium dioxide and from 0 to 20% by weight of aluminum oxide as significant active constituents and silicon dioxide as the remainder, wherein the reaction over the fixed-bed catalyst is carried out continuously in the liquid phase and run at a conversion to oligomerized  $C_n$ -olefins of from 10 to 30% by weight based on a throughput of the reaction mixture through the catalyst in a single pass.

Claim 2: A process as claimed in claim 1, wherein the reaction over the fixed-bed catalyst is run at a conversion to oligomerized  $C_n$ -olefins from 10 to 25% by weight, based on the reaction mixture.

Claim 3: A process as claimed in claim 1, wherein the oligomerization is essentially a dimerization.

Claim 4: A process as claimed in claim 1 carried out at from 30 to 300 °C and a pressure in the range of from 10 to 300 bar.

Claim 6: A process as claimed in claim 1 which is carried out adiabatically in a shaft oven and in which part of the reaction mixture is returned to the reaction.

Claim 7: A process as claimed in claim 1, wherein a feed mixture is fractionated in a column to separate  $C_n$ -olefins and oligomers prior to the reaction, the  $C_n$ -olefins are returned to the reaction, the reaction mixture is returned to the column and the oligomers ( $C_{n+1}$ -hydrocarbons) are discharged.

Claim 8: A process as claimed in claim 1, wherein the reaction mixture after the reaction is fractionated in a column to separate C<sub>n</sub>-olefins and oligomers, the C<sub>n</sub>-olefins are returned to the reaction and the oligomers are discharged.

Claim 9: A process as claimed in claim 1, wherein the reaction mixture is passed over a protective bed wherein catalyst poisons are removed, prior to the reaction.